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LCA ON NANOTECHNOLOGY

Life cycle assessment at nanoscale: review and recommendations

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Abstract
Purpose The need for a systematic evaluation of the human and environmental impacts of engineered nanomaterials (ENMs) has been widely recognized, and a growing body of literature is available endorsing life cycle assessment (LCA) as a valid tool for the same. The purpose of this study is to evaluate how the nano-specific environmental assessments are being done within the existing framework of life cycle inventory and impact assessment and whether these frameworks are valid and/or whether they can be modified for nano-evaluations.
Method In order to do that, we reviewed the state-of-the-art literature on environmental impacts of nanomaterials and life cycle assessment studies on ENMs and nanoproducts. We evaluated the major characteristics and mechanisms under which nanomaterials affect the environment and whether these characteristics and mechanisms can be adequately addressed with current life cycle inventories and impact assessment practices. We also discuss whether the current data and knowledge accumulated around fate, transport, and toxicity of nanomaterials can be used to perform an interim evaluation while more data are being generated.
Observations and recommendations We found that while there is plenty of literature available promoting LCA as a viable tool for ENMs and nanoproducts, there are only a handful of studies where at least some parts of life cycle were evaluated for nanoproducts or nanomaterial. None of the LCA studies on ENMs or nanoproducts that we came across assessed nano-specific fate, transport, and toxicity effects as part of their evaluation citing the lack of data as the primary reason.

However, our literature review indicates that nano-LCA studies need not omit the assessment of nanomaterials' human health and environmental impact due to incomplete data. There is some evidence that scalability may exist in certain types of nanomaterial, and traditional characterization can be applied even below 100 nm up to the scalability breakdown limits. For the size range where the scalability cannot be established, it may be more appropriate to explore empirical relationships, though possibly crude, between nanomaterial properties and their impact on human health and environment. Empirical relationships thus derived can serve as valid input for assessment until specific data points for nanomaterial fate, transport, and toxicity become available. Finally, where there is no quantitative data available, qualitative inferences may be drawn based on the known information of the nanomaterial and its potential release pathways.

Keywords LCA of engineered nanomaterials · LCI of engineered nanomaterials · LCIA of engineered nanomaterials · Nanomaterial fate · Transport · Toxicity

1 Introduction

According to the generally accepted convention, engineered nanomaterials (ENMs) are manufactured material with at least one dimension below 100 nm (Klöpffer et al. 2007). By 2014, these are estimated to capture US $2.6 trillion global market (LUX-Research 2004). The Woodrow Wilson Center's Project on Emerging Nanotechnologies (PEN) estimates that over 1,300 manufacturer-identified, nanotechnology-enabled products have entered the commercial marketplace around the

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world, and the number is expected to reach 3,400 by 2020 (PEN 2011). This number may be in fact very conservative as there is no obligation under the existing legislation for companies to label their products to indicate the use of nanotechnology (Som et al. 2010).

Literature on nano-risk has been growing over the years (Theis et al. 2011). At the heart of this literature is a concern that the ENMs with their size and unique functionality may exhibit unconventional behavior leading to unexpected fate, transport, and toxicity mechanisms in human and other ecological systems. There is also a concern that most research on ENMs thus far has focused on their development and applications in different fields without much consideration to the potential environmental effects throughout their life cycle (Balbus et al. 2006; Maynard et al. 2006; Tervonen et al. 2008; Ray et al. 2009).

To that end, life cycle assessment (LCA) approach has been proposed for a system-level evaluation of ENMs and nanoproducts (Klöpffer et al. 2007; Bauer et al. 2008; Lewinski 2008; Som et al. 2010). Some literature also indicates that LCA can be coupled with comprehensive risk assessment techniques (Davis 2007; Klöpffer et al. 2007; Shatkin 2008; Wardak et al. 2008) in order to evaluate ENMs early on so that informed decisions can be made about the risks and benefits of nanotech and nanoproducts.

This paper will focus on the life cycle assessment studies in the context of engineered nanomaterials. We will first review the properties of engineered nanomaterial that are specific to their nanoscale and are relevant for the fate, transport, and toxicity evaluation. We will then review the existing LCA studies on engineered nanomaterials focusing on whether these studies effectively address the concerns raised regarding the unique characteristics of ENMs. Lastly, we will identify the gaps in information and suggest possible modifications in the LCA approach needed to holistically evaluate the environmental performance of ENMs.

2 Nano-specific properties that affect fate, transport, and toxicity

The properties of nanomaterials are quite different than those exhibited by the corresponding bulk counterparts (Oberdorster et al. 2005; Junam and Lead 2008). For example, gold is inert in bulk form, but when the size is reduced to a few nanometers, gold nanoparticles are very effective oxidation catalysts (Daniel and Astruc 2003; Auffan et al. 2009). The unique properties that make ENMs attractive for various applications also make them susceptible to unique fate, transport, and toxicity phenomena (Oberdörster et al. 2005; Helland et al. 2008), and thus, may need to be treated as new chemical substances (Borm et al. 2006).

Literature on risks of ENMs indicates that a particle's mobility and interaction with ecological systems are influenced not only by its chemical composition, but also by other intrinsic properties like size, shape, functionality as well as by extrinsic factors such as medium of transport and storage and can cause its behavior to deviate from that of its bulk counterpart (Biswas and Wu 2005; Oberdörster et al. 2005; Oberdorster et al. 2005; Nel 2006; Lewinski 2008; Lundqvist et al. 2008; Darlington et al. 2009; Nel et al. 2009; Dhawan and Sharma 2010). At nanoscale, materials of the same chemical composition but different particle-specific intrinsic and extrinsic factors may exhibit different behavior and have different impacts on the environment and on human health. For example, Klaine et al. (2008) showed that fullerenes and carbon nano-tubes (CNTs) behave differently in the environment despite their identical chemical composition (Klaine et al. 2008).

The size of nanoparticle is perhaps the most influential property causing the deviation in a nanomaterial's behavior from that of its bulk counterpart (Dhawan and Sharma 2010). When the size is reduced to the nanoscale, a greater proportion of atoms/molecules become available on the surface of the material compared to that of their bulk counterpart (Poole and Owens 2003). The inverse exponential relationship between particle size and number of molecules at the surface has been established in the literature (Poole and Owens 2003; Oberdorster et al. 2005). As a corollary to the effects of size, the material's morphology may also play a role in its fate and transport (Maynard et al. 2006). Particles in a similar size range may exhibit different behavior regarding solubility, stability, etc. if they are of different morphology (Auffan et al. 2009; Zhou and Keller 2010).

Because the number of atoms or molecules on the surface of the particle may determine the material reactivity, this is key to defining the chemical and biological properties of nanoparticles (Nel 2006). Higher surface-to-volume ratio makes particles more reactive, thus making them more responsive to their surroundings. This alters the particle's tendency to agglomerate and aggregate, thus affecting its mobility as well as stability (Jiang et al. 2008; Auffan et al. 2009; Keller et al. 2010; Oberdörster 2010; Thio et al. 2011).

Auffan et al. (2009) reviewed the size-dependent properties established by Oberdörster et al. (2005) for a variety of inorganic metal and metal oxide nanoparticles. Out of the 51 studies reviewed, 42 had primary particle diameter below 30 nm and showed properties drastically different from those of the corresponding bulk material. The remaining nine studies had particle diameter above 30 nm and did not exhibit any nanoscale-specific properties. According to the authors, at the scale below 30 nm, particles have a size-dependent crystallinity and excess of energy at the surface which makes them unstable, giving them properties different from those of the bulk material.

Just like the intrinsic material properties described above, the extrinsic factors like methods used to manufacture and store also have a significant impact on the behavior of the ENM (Darlington et al. 2009; Oberdörster 2010) via impacting the intrinsic properties. As illustrated in Fig. 1, these factors affect the material properties like particle size, shape, phase, surface chemistry, etc. and also introduce their own range of variation in the properties (Borm et al. 2006; Oberdörster 2010), which in turn affect the behavioral manifestation like solubility, tendency to agglomerate, bioavailability, etc. (Oberdörster 2010). Similarly, the medium in which the particles are released (intentionally, unintentionally via wear-and-tear) also has a significant effect on the fate–transport–toxicity of the particle (Wiesner et al. 2009). For example, an ENM's interaction with natural water components will be different from those with the natural organic matter under a variety of physicochemical conditions such as pH, ionic strength, etc. (Nowack and Bucheli 2007). Besides, although many ENMs are functionalized to get the intended results via altered behavior, changes in functionalization caused by changes in environmental conditions have only been studied marginally so far (Nowack and Bucheli 2007). As a result of many permutations and combinations of the abovementioned factors, the environmental impact of ENM may not be measured by examining just one of these aspects in isolation (Wiesner et al. 2006). Such complexity makes the characterization of fate, transport, and toxicity a challenge (Handy et al. 2008; Meng et al. 2009; Sayes and Warheit 2009; Savolainen et al. 2010), and therefore, toxicity of ENMs has been studied mostly on case-by-case basis (Borm et al. 2006).

3 LCA studies on nanomaterials—a review

According to the ISO standards (ISO-14040 2006), a life cycle covers the extraction of raw materials, manufacturing, use phase, and the end-of-life phase. None of the published LCA studies of ENMs is ISO-compliant since none covers a complete life cycle of engineered nanomaterials or product. Table 1 presents our review of these studies with the focus on how they have handled nano-specific issues.

More than half of the studies limit their focus on the energy and material flows and do not consider the nano-specific fate, transport, and toxicity. While almost all of them cover the manufacturing phase, only three of them cover the use phase. Two out of those three also cover the end-of-life (EOL) phase qualitatively by way of detailing the potential release pathways. For some studies, the use and EOL phases are not considered by the authors. For example, in the study by Lloyd et al. (2005), where the focus is not on a particular product, but on the nanotechnology itself, the use and EOL phases are not discussed. Here, nanotechnology enables the reduced the usage of precious metals as catalysts within the same process delivering the same end product. Similarly, in their screening-level LCA, Meyer et al. (2010) did not consider the end-of-life phase because the nano-silver was assumed to have been washed off during the use phase and hence the EOL was assumed to be the same as for non-nano-silver socks. Likewise, the LCA study by Roes et al. (2007) assumes no difference in the environmental impacts during the use phase between conventional material and nanocomposites and assumes incineration to be the default EOL for both, and hence, does not discuss these two phases.

Wherever the use phase and EOL are discussed, the implications of "nano" are acknowledged as part of the discussion, but not evaluated quantitatively. Consider for example a case study on the use of CNTs in electronic sector by Bauer et al. (2008). Here, the potential risks in the EOL phase have been noted as output in the LCA, but not quantified. Instead a qualitative description of potential release pathways is offered. In the same article, Bauer et al. (2008) evaluate the technique of physical vapor deposition for the three types of nanomaterials, namely, titanium nitride (TiN), titanium aluminum nitride (TiAlN), and TiN + TiAlN. The authors perform it as a cradle-to-gate study. The release of nano-titanium composites is deemed unlikely at the use and EOL phases by the authors since the particles are molten into a solid compound in the product.

The term "nano environmental effects" mostly indicates nano-toxicity (Bauer et al. 2008; Khanna et al. 2008; Meyer

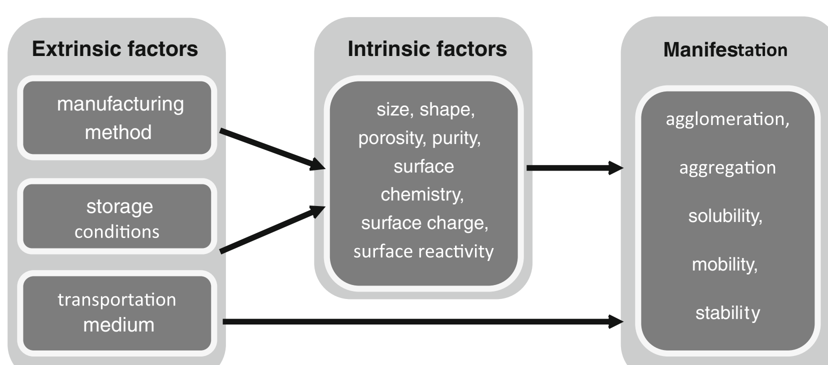


Fig. 1 Summary of relationships between the intrinsic nanomaterial properties, extrinsic factors and behavioral manifestation: A synthesis based on various sources (Borm et al. 2006; Maynard 2006; Nel 2006; Nowack and Bucheli 2007; Klaine et al. 2008; Lewinski et al. 2008; Auffan et al. 2009; Wiesner et al. 2009; Oberdörster 2010)

Table 1 Peer-reviewed LCA studies on ENM

	Reference	ENM type	Focus of study	LC phases				Evaluation of nano-specific environmental effects
				E	M	U	EOL	
1	Lloyd and Lave 2003	Nanoclay-reinforced polymer composites	Replacing auto-body panels made of steel with those of polymer composites and aluminum	O	O	×	×	Toxicity for material composition scenarios based on nano/non-nano options evaluated. Toxicity and manufacturing of the nanocomposite itself is not discussed
2	Steinfeldt et al. 2004 (via Bauer et al. 2008)	Nanoelectronics	Lighting—LEDs	×	×	O	×	
		Nanomaterials/nanoparticles	Chemical/paintings	O	O	×	×	
			Chemical/plastics	O	O	×	×	
			Electronics/displays	O	O	×	×	
3	Lloyd et al. 2005	Nanoscale platinum-group metal (PGM) particles	Evaluating reduction in non-renewable resources like PGM via greater process control offered by nanotech	O	O	×	×	The focus is on how nanotech can help reduce the usage of precious metal within the same process, for the same output as before Toxicity assumed to be less with less usage
4	Osterwalder et al. 2006	Various oxide nanoparticles	Energy comparison of wet and dry synthesis methods for oxide nanoparticle production	×	O	×	×	Release and impact of emissions, associated toxicity (where applicable) during nano-manufacturing and nano-synthesis not considered
5	Roes et al. 2007	Polypropylene Nanocomposite	Compare environmental impacts ad costs with nanocomposite products vis-à-vis those with conventional products	O	O	×	×	Human and ecotoxicity not considered. Use phase assumed to be the same for nano and conventional and incineration for energy was assumed to be the default EOL route
6	Kushnir and Sandén 2008	Fullerenes and CNT	Implications for industrial scale production	O	O	×	×	Focus on projection to the larger-scale production. No discussion on nano-specific environmental impacts
7	Healy et al. 2008	SWCNT	Environmental assessment of SWNT production.	O	O	×	×	Qualitative description of EHS concerns due to nanomaterial in Section 1. No quantified evaluation as part of LCA study
8	Khanna et al. 2008	Carbon nanofibers (CNF)	Environmental burden of CNF synthesis vis-à-vis that by traditional materials	O	O	×	×	Release and impact of CNFs on humans and ecosystem during manufacturing not considered
9	Köhler et al. 2009	Carbon nanotubes (CNT)	Potential release of carbon nanotubes throughout the life cycle of textiles and lithium-ion batteries	×	O	O	O	A study of application-dependent release of CNTs indicating that a release of nanotubes can occur not only in the production phase, but also in the usage and disposal phases of nanotube applications
10	Bauer et al. 2008	TiN, TiAlN, Ti + TiAlN	Examine the implications of life cycle thinking on nanotechnology (and nanoproduct) evaluation; 2 case studies	×	O	×	×	Nano-related emissions not considered as deemed not likely by the authors
		CNT		O	O	O	O	Potential risks at EOL noted, but not quantified
11	Khanna and Bakshi 2009	Carbon polymer nanocomposites	Energy implications for production and use of carbon nanofiber reinforced polymer nanocomposites	O	O	×	×	Production energy requirement comparison
12	Meyer et al. 2010	Silver nano	Identify the life cycle hot spots via screening-level LCA	O	O	×	×	EOL not considered; assumed to be the same as non-nano-silver socks as nano-silver assumed to have been washed off during the use phase
13	Şengül and Theis 2011	QD photovoltaics	LCA	O	O	×	×	Toxicity results not reported
14	Grubb and Bakshi 2011	Titanium dioxide	Evaluate the hydrochloride production process	O	O	×	×	Production energy requirement comparison

ENM engineered nanomaterial, *TiN* titanium nitride, *TiAlN* titanium aluminum nitride, *QD* quantum dot, *SWCNT* single-walled carbon nanotube, *E* extraction, *M* manufacturing, *U* use, *EOL* end-of-life

"O" Indicates that the corresponding LC phase is assessed (either qualitatively or quantitatively) in the study

"×" Indicates that the corresponding LC phase was not assessed in the study

et al. 2009). None of the LCA studies on ENM or nano-products reviewed quantified nanomaterial toxicity, citing the lack of information on the nanomaterial properties as well as that on toxicity and physiochemical characterization models as the reason (see Table 1).

Literature is available recommending supplementing LCAs on ENM with risk analysis, multi-criteria decision analysis, and scenario analysis techniques in order to assess nano-toxicity (Seppälä et al. 2001; Matthews et al. 2002; Linkov et al. 2005; Davis 2007; Linkov et al. 2007; Tervonen and Lahdelma 2007; Shatkin 2008; Seager and Linkov 2009; Wiesner et al. 2009). However, a concrete LCA study on ENMs that utilizes these techniques in practice is currently missing according to our review.

Bauer et al. (2008) propose that the assessment of potential environmental and human health risks may be addressed qualitatively in place of quantification until the latter becomes available. Their case study on CNTs (2008) incorporates this approach where the potential release pathways according to the material size range are documented as part of the assessment. Meyer et al. (2010) employ similar reasoning in their qualitative assessment of release of nano-silver from socks. This approach is developed fully by Köhler et al. (2009). Their paper is based on an in-depth qualitative assessment of application-based CNT release pathways throughout the product lifecycle. Our review did not find any other examples where qualitative assessment of nano-fate, transport, and toxicity was performed.

Another gap apparent in the nano-LCA literature is that of lack of information on the production processes (Osterwalder et al. 2006; Bauer et al. 2008; Khanna et al. 2008; Kushnir and Sandén 2008; Khanna and Bakshi 2009; Grubb and Bakshi 2011). Khanna et al. (2008) caution that the industrial scale nano-LCA results could be gross overestimates as the nano-manufacturing processes are likely to become more efficient with higher yields over time and volume. However, this condition is applicable to all emerging products and technologies and cannot be considered nano-specific.

4 Nano-specific properties and LCA

Populating life cycle inventory (LCI) databases with nanomaterial-related input and output flows of emissions and resources and supplementing them with relevant information like size, shape, and functionalization on nanomaterials are necessary prior to envisaging the characterization of nanomaterial's toxic impacts in life cycle impact assessment (LCIA) (Bauer et al. 2008). At present, the available LCI databases are populated with material and product flows that does not distinguish between the bulk and corresponding nanomaterial, nor do the current characterization methods provide characterization factors for nanomaterials (Bauer et al. 2008). The characterization is in fact linked to the adequacy of nano-specific LCI database in that the former cannot be accomplished successfully unless the databases are expanded to include nano-specific parameters.

The lack of information may not justify the omission of the nanomaterial-specific impact assessment in nanomaterial LCAs. On the other hand, it will take substantial efforts and time until our understanding on nanomaterial behavior and their impact reaches the point that LCI and LCIA can fully address nanomaterial's impact. Hence, it might be useful to devise interim, screening approaches to at least raise a flag for potentially significant issue. One such approach is discussed by Bauer et al. (2008) by developing a qualitative approach proposed by Reijnders (2006) where the initial boundaries can be drawn along the "fixed/embedded" or "free/dispersible" state of the engineered nanomaterial in a specific application. The ENMs in "free" state can be further classified according to their likelihood for dispersion in conjunction with their size range.

A number of sophisticated tools and techniques like USEtox (Rosenbaum et al. 2008), CalTOX (McKone and Enoch 2002), and quantitative structure–activity relationship (QSAR) (Dudek et al. 2006; Puzyn et al. 2009; Puzyn et al. 2010) are available to quantitatively assess the fate, transport, or toxicity of chemicals and bulk material; however, their applicability to nanomaterials, without significant retooling to take into account the intrinsic and extrinsic factors that control ENM behavior, is questionable as indicated by the need for tools and characterization expressed in the most recent literature (Maynard et al. 2011). Modified approaches like quantitative nanostructure–activity relationship (QNAR, which is a modification of QSAR for nano) have been proposed (Fourches et al. 2010) to assess the biological effects of engineered nanoparticles based on the information on their physical, chemical, and geometrical properties. A recent study (Fourches et al. 2011) has demonstrated the application of QNAR via case studies where a preliminary model of QNAR is proposed to predict the activity profile, mainly toxicity, of the manufactured nanomaterials. A study by Subramanian et al. (2009) has highlighted further challenges in the Environmental Health and Safety (EHS) assessment of nanomaterials when nanostructures, like nanoelectromechanical systems, nanomachines, self-healing materials, targeted drugs and chemicals, energy storage devices, and sensor transition from being passive to active with time. The possibility of quantum and synergistic effects due to the matching of scale between nanoparticle and key biological molecules such as proteins and enzymes will likely require case-specific parametric data for the assessment of nanomaterials' EHS impacts (Maynard et al. 2011).

Based on the literature reviewed, we believe that LCA studies on ENM need not omit the assessment of nanomaterials' human health and environmental impact due to

incomplete data. Instead, we propose that the line of thinking illustrated in Fig. 2 be followed in order to perform the assessment at appropriate level. Traditional characterization approaches, where emissions related to bulk material and information on environmental compartments are the bases of assessment, can be followed where scalability is known to exist, for instance in the case of some metal and metal oxide nanomaterial (NM) reviewed by Auffan et al. (2009). In these cases, i.e., where the primary particle size that is currently considered nano (<100 nm) but is technically above the scalability break down in material's behavior, the LCI can be populated for these emissions the same way in which those of their bulk counterpart would be populated, and the same fate–transport and toxicity assessment approaches can be followed in LCIA.

For the size range where the scalability cannot be established, in other words, when the particle size is below the threshold so that bulk material properties cannot be applied, it may be more appropriate to explore empirical relationships, even if simplified, between nanomaterial properties and their impact on human health and environment based on the existing literature, engineering, industrial, and other publicly available data. Empirical relationships thus derived can serve as early input for nano-specific assessments until specific data points for nanomaterial fate, transport, and toxicity become available. Finally, where there is no quantitative data available, qualitative assessment may be performed based on the available information on the nanomaterial and its release pathways. These assessments may follow the technique proposed by Bauer et al. (2008) and Reijnders (2006) as mentioned in the earlier section. Thus, it is possible to assess the environmental impacts of ENM at appropriate levels depending on the type of data available.

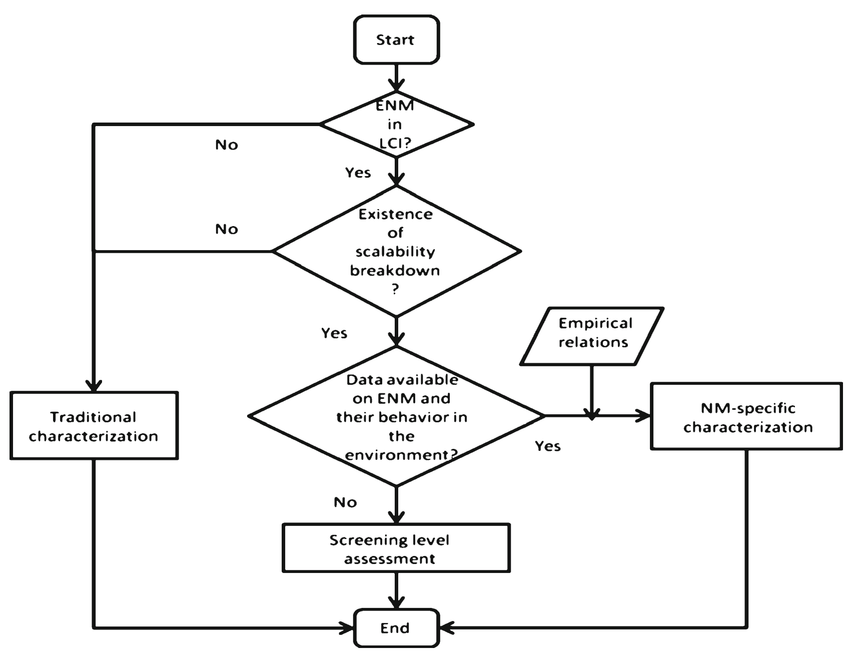
5 Summary and recommendations

Nanotechnology's vast potential is evident from the key role played by its various applications in the UN's Millennium Development Goals (UNESCO 2006). However, embracing nanotechnology without proper risk assessment and risk management can set it on the path of previous cases, like that of CFCs, where commercialization went far ahead of the holistic environmental assessment of these technologies. Nanotechnology's potential benefits have not been properly evaluated against the potential ecological and human health risks, nor is there a holistic assessment of all aspects, not just risks, along the life cycle of nano-based products and services. LCA has already been recognized as a tool able to conduct such a holistic environmental assessment. Accordingly, our research shows that the number of studies on life cycle assessment on ENM is now increasing, though a closer look reveals that a lot of these studies do not cover the entire life cycle of nanomaterials or nanoproducts. Here are our recommendations.

First, populating LCI databases with the ENM-specific information is critically important. Moreover, when compiling LCI, emissions of ENMs should be clearly distinguished from those of their bulk counterpart, and key nanomaterial-specific information including size and shape should be retained as much as possible in order to enable subsequent LCIA for EMNs. Such additional information is particularly important for the emissions beyond the scalability–breakdown point.

Second, while the LCI databases on ENMs keep getting populated for intrinsic properties, protocols and models for their derived properties would be desirable. This is more critical for the use and EOL phases as these

Fig. 2 Flow chart recommending ENM assessment path depending on the availability of data



two phases get frequently omitted in LCAs on ENM due to the lack of modeling techniques available. Developing such protocols and models will enable a holistic assessment that covers the entire life cycle of nano-enabled products.

Third, in the absence of any empirical data, qualitative or screening level assessment can be performed until data become viable. For qualitative assessments, initial boundaries can be drawn based on potential release pathways, material size range, and tendency to disperse, and scenarios can be modeled. This would be a very important step forward toward the incorporation of engineered nanomaterial's environmental assessment as part of its LCA.

Fourth, we recommend incorporating additional information on nanomaterial-specific properties into existing tools like CALtox, USEtox, QSAR, and QNAR to enhance the modeling capability of ENMs' behavior and the impact in the environment. We found only one peer-reviewed case study using QNAR for this purpose (Fourches et al. 2010), and we recognize such an effort as one of the priorities. Moreover, conventional impact categories that are not specific to NMs may well be the most important ones in ENM LCAs, and therefore, such impact categories should receive due attention for ENM impact assessment.

Fifth, we recommend using other relevant tools such as risk assessment in parallel to LCA especially when location-specific parameters are critical in understanding the behavior and impact of an ENM in question. The behavior and impact of ENMs in the environment are relatively more sensitive to the conditions of the environmental compartment to which the ENMs are emitted, and an LCA may not be able to adequately capture such local specifics, which may turn out to be critical.

Finally, although this review is focused on NM-specific issues, it should be noted that LCAs on ENM should not ignore the wisdom from the conventional LCA on bulk materials. As illustrated in Fig. 2, traditional characterization can be implemented for the material that is conventionally classified as "nanomaterial" until the size reaches the point where the scalability breaks down. As the flow chart indicates, at that scale, a systematic ENM-specific characterization can be performed quantitatively, or at least a screening level, qualitative assessment can be performed in order to provide a holistic assessment of engineered nanomaterials and nanoproducts.

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